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Overview on fuel cells

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ABSTRACT

Progress always requires energy. Up today, the energy needs have been provided by combustion of fossil fuels and this has increased the air pollution and the emission of greenhouse gasses. Consequently, the greenhouse emission problem could represents a concrete opportunity to promote the high-efficiency design of conventional plants, but also a new approach to energy systems and the consequent dissemination of advanced technologies. One of the most promising energy conversion technology is the fuel cell because it is an electrochemical device in which the chemical energy is directly converted into electrical energy, with low environmental impact. In this paper the losses of the fuel cells has been analysed.

Contents

1. Introduction

In the course of time, progress has always been associated with the growth of requirement for energy. Up today, the energy needs have been provided by combustion of fossil fuels and this has increased the air pollution and the emission of greenhouse gasses, such as CO_2, in particular in urban area. One of the main problem of the industrialized country is just the management of CO_2 emissions [1–5]. The Kyoto Protocol suggestion of the reduction of the CO_2 emissions can be attained by three primary actions:

1. renewable energy sources;
2. CO_2 sequestration whose estimated investment costs are very high and for this reason it is difficult to be carried out;
3. promotion of existing high efficiency technologies and the adoption of advanced low-CO_2 emission energy systems; indeed, CO_2 reduction is directly related to the thermodynamic efficiency of a plant and an energy policy to promote best existing technologies and their adoption could be developed.

Consequently, the CO_2 emission problem could represents a concrete opportunity to promote the high-efficiency design of conventional plants, the new approach to energy systems and the consequent dissemination of advanced technologies.

In this context, one of the most promising energy conversion technology is the fuel cell [6–19]. In this paper an overview on fuel cell and the thermodynamic approach to irreversibility in fuel cell will be developed. To do so in Section 2 and overview on fuel cell will be summarized, in Section 3 a general thermodynamic approach to fuel cell will be introduced, in Section 4 the irreversibility will be introduced and some numerical evaluation will be presented and in Section 5 some perspectives of fuel cells use will be suggested.

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Table 1
Fuel cells classification.

Characteristics	Polymer electrolyte	Alkaline	Phosphoric acid	Molten carbonate	Solid oxide
Fuel cells					
Operating temperature [°C]	40–80	65–220	205	650	600–1000
Electrolyte	Hydrated polymeric ion exchange membrane	Mobilized or immobilized potassium hydroxide in asbestos matrix	Immobilized liquid phosphoric acid in SiC	Immobilized liquid molten carbonate in $LiAlO_2$	Perovskites (ceramics)
Electrodes	Carbon	Platinum	Carbon	Nickel and nickel oxide	Perovskite and perovskite/metal cermet
Catalyst	Platinum	Platinum	Platinum	Electrode material	Electrode material
Interconnect	Carmon or metal	Metal	Graphite	Stainless steel or nickel	Nickel, ceramic or steel
Charge carrier	H^+	OH^-	H^+	$CO_3{}^+$	O^-

Table 2
Come fuel cells reactions.

Fuel cell kind	Reaction at the anode	Reaction at the cathode
Polymer electrolyte and Phosphoric acid	$H_2 \rightarrow 2H^+ + 2e^-$	$O_2 + 4H^+ + 4e^- \rightarrow 2H_2O$
Alkaline	$H_2 + 2OH^- \rightarrow 2H_2O + 2e^-$	$O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$
Molten carbonate	$H_2 + CO_3{}^{2-} \rightarrow H_2O + CO_2 + 2e$- $CO + CO_3{}^{2-} \rightarrow 2CO_2 + 2e$-	$O_2 + CO_2 + 4e^- \rightarrow 2\ CO_3{}^{2-}$
Solid oxide	$H_2 + O^{2-} \rightarrow H_2O + 2e^-$ $CO + O^{2-} \rightarrow CO_2 + 2e^-$ $CH_4 + 4O^{2-} \rightarrow 2H_2O + CO_2 + 8e^-$	$O_2 + 4e^- \rightarrow 2O^{2-}$

2. The fuel cells

Fuel cell is an electrochemical device in which the chemical energy is directly converted into electrical energy [13,14].

In 1801, the British chemist Humphry Davy (1778–1829) developed his researches on electrolysis using the voltaic pile in order to split up common compounds discovering several new metals, as sodium, potassium and the alkali metals [20]. He laid the scientific foundations for the fuel cells, then designed in 1838 by the German-Swiss chemist Christian Friedrich Schönbein (1799–1868) [21]. In 1839, the Welsh chemical-physicist William Robert Grove (1811–1896) realized the first gas voltaic battery with which he proved that an electrochemical reaction between oxygen and hydrogen produce an electric current [22,23]. The British chemist Ludwig Mond (1839–1909) and Charles Langer used coal as fuel and introduced the term fuel cell obtaining 20 Am^{-2} at 0.73 V [23]. In 1932, the British engineering Francis Bacon modified the Langer's and Mond's cell realizing, in 1958, the first alkaline fuel cell, used later in Apollo spacecraft, and in 1959 he was able to obtain 5 kW system which was really used, with a power of 15 kW, in an agricultural tractor by Harry Karl Ihrig. At the end of the 1950s NASA developed fuel cells for space missions: Willard Thomas Grubb and Leonard Niedrach designed the first Polymer Electrolyte Membrane Fuel Cell, used by NASA in Gemini space programme, while a 1.5 kW Alkali Fuel Cell has been used in Apollo space missions, providing the astronauts both power and drinking water; later, a 12 kW AFC has been used in space shuttle. Since 1970, an interest for use of fuel cells in electric vehicle is growing up today, and since 2007 they are commercialized [24–26].

The core of the fuel cells is the unit cell, the component in which the device converts the chemical energy in electrical energy. It consists of an electrolyte in contact with an anode (negative electrode) and a cathode (positive electrode). The fuel cells are classified in relation to their electrolytes and fuels used as follows [24,25]:

1. PEMFC (proton exchange membrane or polymer electrolyte membrane fuel cell) uses a water-based, acidic polymer membrane as the electrolyte and platinum-catalysed electrodes. It uses pure hydrogen, but also reformed natural gas, removing carbon monoxide. Its operative temperature is below 100 °C;
2. HT-PEMFC (high temperature PEMFC) is a PEMFC obtained by changing the electrolyte from a water-based to a mineral acid-based system. It operates up to 200 °C;
3. DMFC (direct methanol fuel cell) uses a polymer membrane as the electrolyte and the platinum–ruthenium catalyst on its anode uses the hydrogen from liquid methanol directly;
4. MCFC (molten carbonate fuel cell) uses a molten carbonate salt suspended in a porous ceramic matrix as the electrolyte with coal-derived fuel gas, methane or natural gas, operating at temperatures of about 650 °C;
5. PAFC (phosphoric acid fuel cell) consists of an anode and a cathode made of a finely dispersed platinum;
6. catalyst on carbon and a silicon carbide structure that holds the phosphoric acid electrolyte. It operates up to 200 °C;
7. SOFC (solid oxide fuel cell) uses a solid ceramic electrolyte;
8. AFC (alkaline fuel cell) uses an alkaline electrolyte and is fuelled with pure hydrogen and oxygen.

Their properties are summarized in Table 1 while some fuel cells reactions are summarized in Table 2 [28–36]. Today, fuel cells can be applied in power systems as follows [26,27]:

1. in the rang 1 W–10 kW: cell phones, personal computer and personal electric equipments;
2. in the range 1–100 kW: power vehicles and public transportation;
3. 1–10 MW: power systems for energy.

3. Thermodynamic analysis of fuel cells

The reversible cell potential is the maximum electrical energy potential which can be obtained in a fuel cell. Of course, also in fuel cells the irreversibility occurs. The aim of this Section is to analyze it.

To do so, it is fundamental to define the system considered. It is the volume in which fuel (H_2 for example) and oxidan (O_2 for example) flows enter and the products (H_2O if H_2 and O_2 are considered) of the chemical reaction exit. The fuel cell is dipped in a thermal bath at constant temperature. The reactants and the reaction products flows have the same temperature T and pressure p [37]. The system considered is at the steady state. The chemical reaction can be represented by the following general chemical equation:

$$\text{Fuel}+\text{Oxidant}\rightarrow\text{Product}+\dot{Q}+\dot{W}$$

$$\sum_{rc}N_{rc}\text{ reactants}\rightarrow\sum_{pr}N_{pr}\text{products} \tag{1}$$

where $\dot{Q}$ is the heat power produced and $\dot{W}$ is the power done, rc means reactants and pr products. For the system considered the First Law of thermodynamics can be written as [38–41]

$$\tilde{G}(\tilde{h}_{in}-\tilde{h}_{out})+\dot{Q}+\dot{W}=0 \tag{2}$$

and the Second Law results [38–41]

$$\frac{\dot{Q}}{T}+\tilde{G}(\tilde{s}_{in}-\tilde{s}_{out})+\dot{S}_g=0 \tag{3}$$

where $\tilde{G}$ is the molar flow, $\tilde{h}$ is the molar enthalpy, $\tilde{s}$ is the molar entropy and $\dot{S}_g$ is the entropy generation rate

$$\tilde{h}_{in}=\tilde{h}_{rc}=\frac{1}{N_f}\sum_{rc}N_{rc}\tilde{h}_{rc}=\tilde{h}_f+\frac{\tilde{G}_{ox}}{\tilde{G}_f}\tilde{h}_{ox} \tag{4}$$

$$\tilde{h}_{out}=\tilde{h}_{pr}=\frac{1}{N_f}\sum_{pr}N_{pr}\tilde{h}_{pr}=\frac{\tilde{G}_{pr}}{\tilde{G}_f}\tilde{h}_{pr} \tag{5}$$

$$\tilde{s}=\tilde{s}_{rc}=\frac{1}{N_f}\sum_{rc}N_{rc}\tilde{s}_{rc}=\tilde{s}_f+\frac{\tilde{G}_{ox}}{\tilde{G}_f}\tilde{s}_{ox} \tag{6}$$

$$\tilde{s}_{out}=\tilde{s}_{pr}=\frac{1}{N_f}\sum_{pr}N_{pr}\tilde{s}_{pr}=\frac{\tilde{G}_{pr}}{\tilde{G}_f}\tilde{s}_{pr} \tag{7}$$

while f means fuel, ox oxidant, pr products, in inflow and out outflow.

Now, considering the relations (2) and (3), dividing it for the fuel molar flow $\tilde{G}$, it is possible to obtain:

$$\tilde{q}=T(\tilde{s}_{out}-\tilde{s}_{in})+T\tilde{s}_g \tag{8}$$

and

$$\tilde{w}=\tilde{g}_{in}-\tilde{g}_{out}+T\tilde{s}_g \tag{9}$$

with $\tilde{w}=\dot{W}/\tilde{G}$ specific molar work, $\tilde{s}_g=\dot{S}_g/\tilde{G}$ molar entropy generation and $\tilde{g}=\tilde{h}-T\tilde{s}$ Gibbs molar potential, function of the temperature and the pressure of the system, $\tilde{q}=\dot{Q}/\tilde{G}$ molar specific heat [38–41].

The maximum power can be obtained in reversible processes, when $\tilde{s}_g=0\,\text{J}\,\text{K}^{-1}\,\text{mol}^{-1}$, and it results $\dot{W}=\tilde{G}(\tilde{g}_{in}-\tilde{g}_{out})$. This maximum power can be expressed as a function of electromotive force, known as cell potential, E which can be written as

$$E=\frac{\tilde{w}}{nF}=\frac{\tilde{g}_{in}-\tilde{g}_{out}}{nF}-\frac{T\tilde{s}_g}{nF}=E_{\max}-E_\lambda \tag{10}$$

with n number of moles of electrons and F Faraday constant, E_{max} the maximum cell potential in reversible conditions and E_λ the voltage lost (not obtained) due to irreversibility

$$E_\lambda=\frac{T}{nF}\tilde{s}_g \tag{11}$$

From the definition of cell potential (10), its change in relation to temperature results [37]

$$\left(\frac{\partial E}{\partial T}\right)_p=-\frac{1}{nF}\left(\frac{\partial(\Delta\tilde{g})}{\partial T}\right)_p \tag{12}$$

and in relation to pressure results

$$\left(\frac{\partial E}{\partial p}\right)_T=-\frac{1}{nF}\left(\frac{\partial(\Delta\tilde{g})}{\partial p}\right)_T \tag{13}$$

Now, considering the definitions of molar specific Gibbs function

$$\tilde{g}=\tilde{h}-T\tilde{s} \tag{14}$$

and of molar specific enthalpy

$$\tilde{h}=\tilde{u}+p\tilde{v} \tag{15}$$

with $\tilde{u}$ molar specific internal energy and $\tilde{v}$ molar specific volume, it is possible to obtain the following relation

$$d\tilde{g}=\tilde{v}dp-\tilde{s}dT \tag{16}$$

Considering these last relations, in relation to the variation of the molar specific Gibbs function $\Delta\tilde{g}$ between the products and the reactants, it is possible to obtain the variations of the molar specific entropy $\Delta\tilde{s}$ and volume $\Delta\tilde{v}$ between the products and the reactants, too

$$\left(\frac{\partial(\Delta\tilde{g})}{\partial T}\right)_p=-\Delta\tilde{s} \tag{17}$$

$$\left(\frac{\partial(\Delta\tilde{g})}{\partial p}\right)_T=\Delta\tilde{v} \tag{18}$$

Finally, it follows:

$$\left(\frac{\partial E}{\partial T}\right)_p=-\frac{1}{nF}\Delta\tilde{s}(T,p) \tag{19}$$

and in relation to pressure results

$$\left(\frac{\partial E}{\partial p}\right)_T=-\frac{1}{nF}\Delta\tilde{v} \tag{20}$$

From these results, considering that for a great number of a reversible fuel cells the molar specific entropy variation is negative, it is possible to argue [37]:

1. if $\Delta\tilde{s}>0$ for $\Delta N=N_{pr}-N_{rc}>0$, with N_{pr} moles number of products and N_{rc} moles number of reactants, E increases with temperature and decreases with pressure;
2. if $\Delta\tilde{s}=0$ for $\Delta N=0$, E is independent of temperature and temperature;
3. if $\Delta\tilde{s}<0$ for $\Delta N<0$, E decreases with temperature and increases with pressure.

These results are based on pure reactants and products, but in real fuel cells fuel and oxidants flows are diluted by many other species [37]. Consequently, fuel cell potential is affected by the effect of inert diluents, which increase the mass transport resistance, so that the molar specific change of the fuel, oxidant and products must be evaluated at their partial pressure at the mixture temperature.

During the chemical reaction, the heat lost can be evaluated using the second law (3) per unit molar flow of fuel

$$\tilde{q}=T(\Delta\tilde{s}-\tilde{s}_g) \tag{21}$$

while the useful work results

$$\tilde{w} = -(\Delta\tilde{g} + T\tilde{s}_g) \tag{22}$$

Consequently, the first law efficiency [42,43] can be obtained as

$$\eta = \frac{\tilde{w}}{-\Delta\tilde{h}} = \frac{\Delta\tilde{g} + T\tilde{s}_g}{\Delta\tilde{h}} \tag{23}$$

and its maximum value can be obtained in the reversible cell for which $\tilde{s}_g = 0\ \mathrm{J\,K^{-1}\,mol^{-1}}$, so

$$\eta = \frac{\Delta\tilde{g}}{\Delta\tilde{h}} \tag{24}$$

As Li [37] pointed out, a reversible fuel cell has the same maximum efficiency of a Carnot ideal engine [42,43]; indeed, considering an ideal engine which works between the high temperature T_1 and the lower temperature T_2, absorbing the molar heat $\tilde{q}_1$ from the thermostat at temperature T_1, and changing the molar heat $\tilde{q}_2$ with the thermostat at temperature T_2, the first law per unit mole flow of fuel can be written as [38–41]

$$\tilde{w} = \tilde{q}_1 - \tilde{q}_2 \tag{25}$$

with

$$\tilde{q}_1 = -\Delta\tilde{h} \tag{26}$$

and the second law per unit molar flow of fuel [38–41]

$$\tilde{s}_g = \Delta\tilde{s} + \frac{\tilde{q}_1}{T_1} \tag{27}$$

recalling the definition of molar Gibbs function, the Carnot efficiency for such ideal engine results [37,42,43]:

$$\eta_{max} = 1 - \frac{T_2}{T_1} = \frac{T_1 - T_2}{T_1} = \frac{(T_1 - T_2)\Delta\tilde{s}}{T_1\Delta\tilde{s}} = \frac{\Delta\tilde{h} - T_2\Delta\tilde{s}}{\Delta\tilde{h}} = \frac{\Delta\tilde{g}}{\Delta\tilde{h}} = \frac{\Delta\tilde{g}}{\Delta\tilde{g} + T\Delta\tilde{s}} = \frac{E}{E - T(\partial E/\partial T)_p} \tag{28}$$

as Li underlined [37].

4. Irreversibility evaluation in fuel cells

In reversible conditions, the energy loss is the heat lost towards the environment $T\Delta\tilde{s}$, due to the negative entropy. To this reversible loss the work produced when the fuel cell supplies an electric current I:

$$\dot{W} = EI \tag{29}$$

In this case, the cell potential, and its related efficiency, decreases in increasing of current [37] as a consequence of the following phenomena:

1. activation polarization E_λ^{act}, due to the irreversibility of the electrochemical reactions and evaluated by the Tafel relation $E_\lambda^{act} = a + b\ \ln I$, with $a = -RT/(\alpha nF)$, being α the electron transfer coefficient, $b = -a$ called the Tafel slope obtained by the plot of E_λ^{act} as a function of I;
2. ohmic polarization $E_\lambda^{ohm} = rI$, due to electrical resistance r in the fuel cell. The resistance r is the total resistance of the fuel cell, sum of the electronic, R_{el}, ionic R_{ion} and contact, R_{conct}, effects: $r = R_{el} + R_{ion} + R_{conct}$;
3. concentration polarization E_λ^{conc}, due the overaccumulation of products in the reaction area, expressed as $E_\lambda^{conc} = b\ \ln(1 - I/I_L)$, with I_L the limiting current, a measure of the maximum rate at which a reactant can be supplied to an electrode;
4. Nernst loss $E_\lambda^{Nernst} = (RT/nF)\ln(k_{out}/k_{in})$, with R universal constant of gas, k equilibrium constant for partial pressure evaluated for the inlet and outlet gas composition, due to the spontaneous adjustment of the lowest electrode potential by the cell.

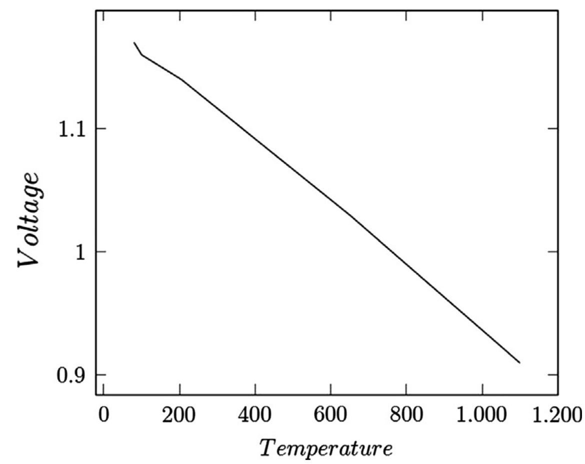


Fig. 1. Cell reversible potential (Voltage [V]) vs temperature [°C].

The resultant loss can be obtained by the following relation

$$E_\lambda = E_\lambda^{act} + E_\lambda^{ohm} + E_\lambda^{conc} + E_\lambda^{Nernst} \tag{30}$$

from which the entropy generation [39–50] results

$$\tilde{s}_g = \frac{E_\lambda}{nFT} \tag{31}$$

and the related heat lost (21) for irreversibility can be evaluated as

$$\tilde{q} = T\Delta\tilde{s} - nFE_\lambda = \Delta\tilde{h} - \Delta\tilde{g} - nFE_\lambda \tag{32}$$

while the equivalent power loss results

$$\dot{w} = I\frac{\tilde{q}}{nF} = I\left(\frac{\Delta\tilde{h} - \Delta\tilde{g}}{nF} - E_\lambda\right) \tag{33}$$

Now it is interesting to obtain numerical evaluation on the previous analytical relations. Considering the hydrogen oxidation, represented by the chemical reaction

$$2H_2 + O_2 \rightarrow 2H_2O \tag{34}$$

For this reaction the standard (temperature = 25 °C and pressure = 1 atm) molar specific enthalpy variation $\Delta\tilde{h}$ is $-286.0\ \mathrm{J\,mol^{-1}}$ while the standard molar specific Gibbs function $\Delta\tilde{g}$ is $-237.3\ \mathrm{J\,mol^{-1}}$. At this condition, the reversible potential E_{max} results 1.23 V, with an ideal efficiency of 83.0%. Using the relation (12) it is possible to evaluate the effect of temperature on this ideal potential: the result is represented in Fig. 1. The loss are evaluated and represented in Fig. 2.

Now, let us consider a PEMFC for car use. It involves the C_2H_5OH and H_2O to produce hydrogen, with a reaction efficiency around 96%. No pollutants or poisons are produced. The theoretical efficiency for hydrogen fuel cell has been estimated around the 60% [51–53]. Considering the previous thermodynamic relations it is possible to evaluate the real efficiency as a rate of the produced voltage ($E = 0.75$ V) on the voltage corresponding to the hydrogen high heating value ($\Delta H^\circ = -285.9\ \mathrm{kJ\,mol^{-1}}$ corresponding to $E = 1.48$ V), obtaining an efficiency of 50%, which, considering the losses, becomes 38%, in accordance with the range evaluated by Fleischer and Ørtel of 31–39% [24].

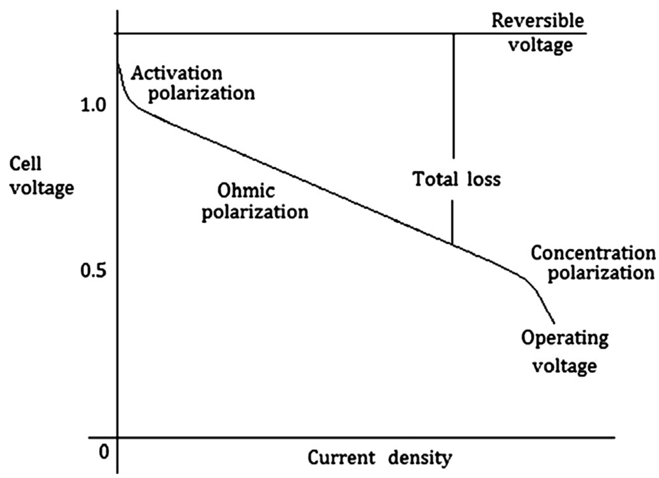


Fig. 2. Cell voltage [V] vs current density [mA m^{-2}].

5. New perspective for fuel cell use

In relation to the reports on production [24], in 2010 fuel cells grew by 40% in market, of which the 95% is accounted to portable fuel cell, while the 97% of fuel cells used PEMFC technology. This important goal can be explained on relation to the versatile application of fuel cell technology; indeed, it can replace a range of power supplies from batteries to internal combustion engines from home heating to mobile phone chargers and cars. Moreover, fuel cells can use any fuel as a source of hydrogen. This can offer great possibilities for the technology, from integrating directly into existing natural gas infrastructure to processing waste hydrogen and using bio-methane. In particular, the portable sector is the largest market with growth of micro-portable fuel cells, representing only the 2.6% of the global power supplied.

Fuel cell can also be designed as auxiliary power units (APU) in order to provide hotel loads of up to 100 W or integrated into vehicles. The APU market is consider a growing field of production because fuel cells offer a low-emission and low-noise technology. Indeed, when methane is used as fuel, the fuel cells can produce up to 8 Nm^3 of hydrogen per hour, providing electricity and heat during the day and hydrogen overnight to be used to fuel buses or other vehicles.

Another use is in the provision of low oxygen-content air for fire suppression systems, because the exhaust gas contains 15% oxygen, too low to support combustion.

6. Conclusions

Fuel cells are electrochemical devices which convert chemical to electrical energy [53–62]. They are realized by an electrolyte medium [34,35] between two electrodes, of which the anode allows the oxidation while the cathode the electrochemical reduction. The oxidation–reduction chemical reaction produces ions and electrons: the electrons produced at the anode represents the external electric current.

Fuel cell can represent an actual power generation technology with a limited environmental impact. In order to use them, it is necessary to evaluate their second law efficiency and in particular their losses. To do so a thermodynamic approach to fuel cells and the analysis of their losses have been developed. The result consists in thermodynamic and thermochemical relation which allow to evaluate the losses which are:

1. the mixed potential at electrodes, generated by chemical reactions at the electrodes;
2. activation losses, due to kinetics at the electrodes;
3. ohmic losses, generated by the resistive losses in the electrolyte and in the electrodes;
4. mass transport losses, due to non reacting diffusion in the gas-diffusion layer and to reacting diffusion at the electrode layers.

The relations obtained have been evaluated, obtaining results in accordance with the literature [63,64].

Even if there exist losses and dissipation, the fuel cells have a high efficiency and they can really represent a real technology for power use with low environmental impact in a next future [65–68].

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